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Elastic properties of terbium

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The temperature dependence of the Young modulus along the crystallographic axes b and c (E_b and E_c), and the internal friction of a terbium single crystal have been measured. At 4.2 K, E_b and E_c are equal to 38 and 84.5 GPa, respectively. The lattice part of the Young modulus and the Debye temperature has been calculated. The origin of the Young modulus anomalies arising at the transition to the magnetically ordered state is discussed. [S0163-1829(96)00729-1]

The rare-earth metal terbium has a hexagonal structure, and a magnetocrystalline anisotropy characteristically for heavy rare-earth metals with nonzero orbital moments.[1] Terbium undergoes two magnetic phase transitions: from paramagnetism to a helical spin structure at Θ_N, and from the helical spin structure to a ferromagnetic state at Θ_c.[2–4] An external magnetic field destroys the helical spin structure at a critical value H_{cr} of the field. According to various investigations, the value of Θ_c lies in the range of 210–220 K, Θ_N in the range of 223.3–231 K, and $H_{\text{cr}}=100$–1000 Oe.[5–7]

The Young modulus (E), elastic constants (c_{ij}), ultrasonic attenuation, and internal friction (Q^{-1}) have been investigated on polycrystalline and single-crystal samples of terbium.[6–16] Anomalies connected with the change of magnetic order were found in the temperature dependence of E and c_{ij} at the phase transitions. Most of the studies were done with the help of ultrasonic methods at frequencies about 10 Mhz. Due to the relaxation character of the Young modulus and the internal friction at such a high frequency, important information can be lost. At lower frequencies measurements were done on a single crystal above the liquid-nitrogen temperature.[13,14]

In this work, the measurements of the Young modules and the internal friction of the terbium single crystals were made in the temperature range 4.2–390 K and at the frequency 1.5 kHz. The Young modulus (E) and internal friction (Q^{-1}) of the terbium single crystals were measured along the crystallographic c and b axes. The elastic properties were determined by the method of flexural autovibrations of a cantilevered thin rod as described in Ref. 17.

The sample purity was 99.9 at. %. The crystal was oriented using a diffractometer. The sample was cut by the electrospark method to the rod with dimensions of $7\times2\times0.2$ mm^3. After cutting, the sample was etched in a HNO_3-C_2H_5OH solution in order to remove the destroyed layer. The long edge of the sample was parallel to the crystallographic b axis for E_b measurements, and parallel to the c axis for E_c measurements.

The temperature dependence of the Young modulus and the internal friction measured along the crystallographic c and b axes are shown in Figs. 1 and 2, respectively. In Fig. 1, E_c shows a general trend to decrease with heating, displaying minima in the vicinity of the magnetic phase transitions. The phase-transition temperatures were determined from the positions of the minima on $E_c(T)$ as $\Theta_c=220$ K and $\Theta_N=229$ K. At 4.2 K, E_c is equal to 84.5 GPa. In the temperature dependence of Q^{-1} along the c axis (Fig. 1) there are two maxima corresponding to Θ_c and Θ_N. The temperature dependence of $E_b(T)$ as shown in Fig. 2 differs significantly from that of $E_c(T)$. The Young modulus decreases upon heating from 4.2 until about 150 K, and in the region of Θ_c and Θ_N a sharp increase of E_b is observed. In the paramagnetic phase, E_b decreases monotonously with heating. At $T=228$ K there is an anomaly which can be connected with the transition at Θ_N. The temperature Θ_c was determined as the point of the most rapid change in E_b to be 220 K.

Take notice of the considerable rise in internal friction in the low-temperature region, which was observed in measurements along the b axis (Fig. 2). Earlier, a significant low-

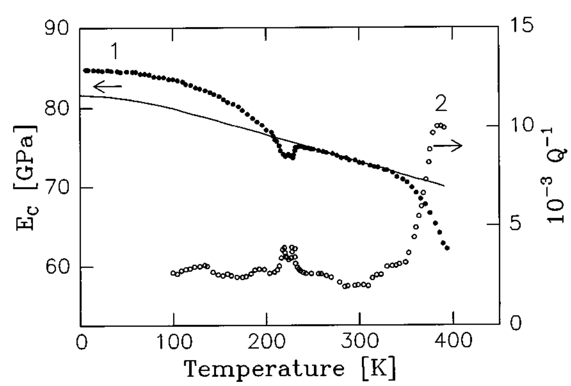

FIG. 1. Temperature dependence of the Young modulus E_c (curve 1) and internal friction Q^{-1} (curve 2) measured along the crystallographic axis c. The lattice part of E_c is shown by the solid line.

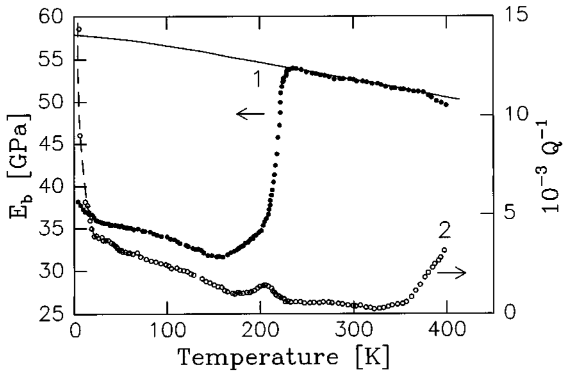

FIG. 2. Temperature dependence of the Young modulus E_b (curve 1) and internal friction Q^{-1} (curve 2) measured along the crystallographic axis b. The lattice part of E_b is shown by the solid line.

temperature value of the internal friction was found in Dy,[18] Er, and Gd-Dy alloys.[19,20] In Ref. 21, this was explained by the energy loss due to motion of narrow domain walls with high intrinsic coercivity caused by alternative mechanical strains. The magnetic part of the Young modulus is calculated as $E^{(m)}(T)=E_{\mathrm{exp}}(T)-E_l(T)$, where $E_{\mathrm{exp}}(T)$ is the above experimentally determined value of the Young modulus. The lattice part of the Young modulus is shown in Figs. 1 and 2 by solid lines. This shows the applicability of the model[22–26] for the analysis of the temperature dependence of $E(T)$ with the average Debye temperature $\Theta_D=175$ K. The calculated values of the lattice part of the Young modulus at $T=0$ K are as follows: for the b axis $E_0=57.65$ GPa; for the c axis $E_0=81.5$ GPa. In general, the curve for $E^{(m)}(T)$ repeats the behavior of $E_c(T)$ and $E_b(T)$. If we do not take into account the domain structure of the sample, the main reasons for the appearance of the Young modulus anomaly at the transition to the magnetically ordered state are the following.

(i) Additional magnetostriction deformations of the sample caused by the change in spontaneous magnetization under the influence of mechanical stresses acting on the sample during the measurement.

(ii) The change of crystalline lattice stiffness caused by spontaneous (or forced) magnetostriction in the magnetic phase. The magnetostriction arising under magnetization leads to the establishment of modified interatomic distances with different values of bounding forces between the atoms.

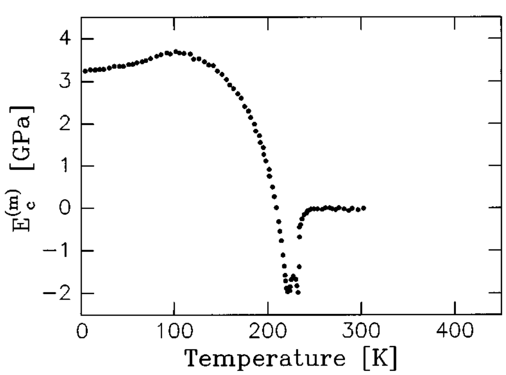

FIG. 3. Temperature dependence of the magnetic part $E_c^{(m)}$ of the Young modulus E_c.

The energy consideration (i) is consistent with a negative contribution to the anomaly of the Young modules.[25] The mechanostriction contribution is high only in the vicinity of the magnetic phase transition where the spins are weakly coupled, and the influence of mechanical stress on magnetization is large. In the low-temperature region, the coupling between the spins becomes stronger, and the mechanostriction contribution to the Young modulus anomaly becomes large. So at low temperatures, the Young modulus anomaly cannot be explained by the mechanostriction mechanism. In addition, in the low-temperature region in terbium a positive magnetic contribution $E_c^{(m)}$ is observed.

In Ref. 26, the thermodynamic theory of second-order phase transitions taking into account the relaxation process was used for the description of the Young modulus anomaly at the Curie point of an isotropic ferromagnetic single domain. The following equation was obtained for the Young modulus anomaly caused by the mechanostriction of the paraprocess:[26]

$$\frac{\Delta E}{E_l}=\frac{-\Delta_E E_l}{1+\omega^2\tau^2}, \tag{1}$$

$$\Delta_E=\frac{\gamma^2 I^2}{\frac{H}{I}+2\beta I^2}, \tag{2}$$

$$\tau=\frac{1}{k\left(\frac{H}{I}+2\beta I^2\right)}, \tag{3}$$

where $\Delta E=E-E_l$ is the Young modulus anomaly at the Curie point, τ is the relaxation time, Δ_E is the relaxation degree of the Young modulus, I is the magnetization of the sample, H is the magnetic field, ω is the frequency of the sample oscillations, γ and β are the thermodynamic coefficients, and k is the kinetic coefficient characterizing the velocity of approaching the equilibrium magnetic state. The thermodynamic coefficient of magnetostriction γ can be determined experimentally from the field dependency of the volume magnetostriction in the range of transition to the magnetically ordered state. The value of $\gamma=1.8\times10^{-9}$ G^{-2} was obtained from an analysis of the data of Ref. 27, where the field dependency of the volume magnetostriction of terbium along the b axis was measured. Measurements of terbium magnetization along the b axis[30] made it possible to determine $\beta=2\times10^{-5}$ G^{-2}. Using these values of γ and β, and using $E_l=53.5$ GPa at Θ_N, we have calculated $\Delta E_b=-2.3$ GPa. From Fig. 4 one can see that the anomaly of E_b observed in the magnetically ordered state is considerably higher. Thus this anomaly cannot be explained by the mechanostriction mechanism only. However, the model proposed in Ref. 26 was worked out for a ferromagnet, but below Θ_N terbium is a helical antiferromagnet. Thus, in the general case, it is necessary to take into account the influence of the elastic stress not only on the ferromagnetic but also on the helical spin structure. Furthermore, the existence of domain structures must be considered. In Ref. 28, it was established that the unmagnetized terbium consists of plate-type domains separated by 180° domain walls lying in

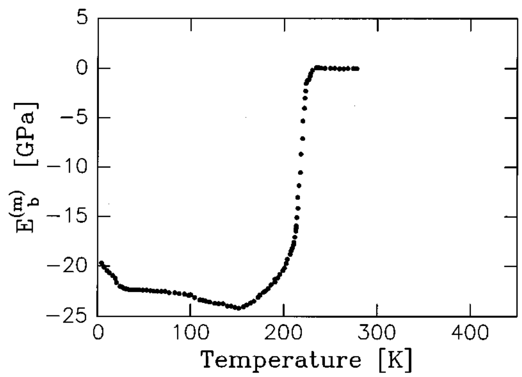

FIG. 4. Temperature dependence of the magnetic part $E_b^{(m)}$ of the Young modulus E_b.

the basal plane. Such a structure must give a minimal mechanostriction contribution to the change of the Young modulus E_b in the magnetically ordered state. This is confirmed by our measurements of E_b in a weak magnetic field aligned along the b axis, and by the data of Ref. 7. In the ferromagnetic phase in the field where the domains can already exist, the change of E_b caused by the field is negative, and equals 3% of the Young modulus anomaly arising in the magnetically ordered state.

The third reason is probably the mechanism mentioned above, connected with the crystalline lattice stiffness change caused by the magnetostriction. The model proposed in Ref. 26 was developed in Ref. 29, where the change of crystalline lattice stiffness was taken into account. As a result, a more general equation than Eq. (1) was derived for the Young modulus anomaly at the Curie point:

$$\frac{\Delta E}{E_l} = \frac{-\Delta_E E_l}{1+\omega^2\tau^2} + \varepsilon I^2 E_l, \tag{4}$$

or, for $H=0$ and $\omega=0$,

$$\frac{\Delta E}{E_l} = \frac{-\gamma^2 E_l}{2\beta} + \varepsilon I_s^2 E_l, \tag{5}$$

where ε is the thermodynamic coefficient, and I_s the spontaneous magnetization.

The second term in Eq. (4) is due to the stiffness change. On the contrary, it is the first mechanostriction term which does not have a relaxation character and does not depend on the frequency. The contribution from the change of stiffness will grow in the low temperature region because of rising I_s.

It can be assumed that the E_b anomaly in terbium is connected with a change of the stiffness of the crystalline lattice. Apparently, this is also the cause of E_c rising upon cooling. The significant difference in the temperature dependency of the Young modulus magnetic parts $E_c^{(m)}$ and $E_b^{(m)}$ (the negative sign of $E_b^{(m)}$ and the positive sign of $E_c^{(m)}$ in the low-temperature region) can be explained by the strong anisotropy of the coefficient ε and its different sign for the b and c axes.

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